

BASIS FOR THE AMENDMENT

Claims 1-33 are active in the present application. Claims are new claims. Support for new Claims 32 and 33 is found on page 36 and on page 34, line 36 to page 35, line 23.

Support for new Claims 30 and 31 is found on page 6, lines 27-32 and on page 36. The claims have been amended for matters of form and to remove multiple dependencies. The specification has been amended to correct a typographical error. Support for the amendment to the specification is found in the original claims. No new matter is added.

REQUEST FOR RECONSIDERATION

Applicants thank Examiner Johnson for the helpful and courteous discussion of July 7, 2005.

Applicants have described an invention that includes a multimetal oxide material of the formula (I) recited in present independent Claim 1. The claimed multimetal oxide contains vanadium (V), molybdenum (Mo) and oxygen, in addition to at least three other elements identified as  $M^1$ ,  $M^2$  and  $M^3$ .  $M^1$ ,  $M^2$  and  $M^3$  are different from one another and different from Mo and V. In the specification as originally filed Applicants noted that other multimetal oxide materials containing Mo and/or V are known. The deficiencies of the prior art multimetal oxide materials are explicitly discussed, see for example page 4, lines 21-29 where it is stated:

What is disadvantageous about the stated prior art, however, is that, on the one hand, it does not answer the question as to whether the promoters are incorporated both into the i phase and into the k phase and whether they influence the catalytic activity of both phases and, on the other hand, their multimetal oxide materials are not completely satisfactory, with regard to the selectivity of the formation of desired product, as catalysts for the heterogeneously catalyzed partial oxidation and/or ammoxidation of saturated and/or unsaturated hydrocarbons.

Thus not only are the multimetal oxide materials of the present claims defined by their compositional characteristics (e.g., they are “five-metal” catalysts), they are also defined by their crystal phase. The crystal phase of a multimetal oxide may be a k-phase and/or an i-phase. The i-phase may be distinguished from the k-phase by the x-ray diffraction spectra wherein the i-phase has no reflection at the  $2\theta$  at  $50.0 \pm 0.3^\circ$  (page 3, lines 5-9).

Conventional catalysts may contain a mixture of the i-phase and k-phase or be of pure k-phase. A multimetal oxide material that is rich in the i-phase relative to the k-phase may be prepared by removing the k-phase by differential solubility (paragraph bridging pages 7 and 8).

In contrast to the processes which are conventionally used to prepare multimetal oxides and result in the formation of both a k-phase and an i-phase, the multimetal oxide of the invention is prepared in a manner that yields a relatively greater quantity of the i-phase. This is reflected in the claimed invention which recites multimetal oxide material “whose x-ray diffraction pattern has no reflection with the peak i position  $2\theta = 50.0 \pm 0.3^\circ$ ” (see last two lines of Claim 1). The invention multimetal oxide material must be one that is enriched in the i-phase relative to the k-phase. This requirement is present in independent Claim 1 by the requirement that the reflection of a peak of the i-phase relative to the reflections of the i- and k-phases is from 0.65 to 0.85. Therefore, the claimed multimetal oxide material is one that contains a specific i-phase multimetal oxide and wherein the i-phase multimetal oxide is present in a greater quantity than the k-phase multimetal oxide as required by the ratio of the i-phase reflectance over the total reflectance of the i- and k-phases.

In order to demonstrate the advantages of the invention multimetal oxide, Applicants prepared a number of Inventive and Comparative Examples. The results of Applicants' experiments are tabulated on page 36 of the specification. The Table provides the compositional formulas for a number of multimetal oxides as well as their effectiveness in the selective catalytic formation of acrylic acid (see column labeled  $S_{ACA}$ ) and their effectiveness in selective propene byproduct formation (see the column labeled  $S_{PEN}$ ). The total propane conversion is provided in the column labeled  $C_{PAN}$ .

In the Table shown below the Inventive Examples are prefixed with “E” and the comparative Examples are prefixed with “CE”.

Table

Example	Composition	R	T [°C]	C <sub>PAN</sub> (mol%)	S <sub>ACA</sub> (mol%)	S <sub>PEN</sub> (mol%)
CE1	Mo <sub>1</sub> V <sub>0.33</sub> Te <sub>0.22</sub> Nb <sub>0.11</sub> Ni <sub>0.01</sub>	0.26	390	30	66	9
E1	Mo <sub>1</sub> V <sub>0.29</sub> Te <sub>0.14</sub> Nb <sub>0.13</sub> Ni <sub>0.007</sub>	0.71	390	80	66	2
CE2	Mo <sub>1</sub> V <sub>0.33</sub> Te <sub>0.19</sub> Nb <sub>0.11</sub> Pd <sub>0.01</sub>	0.25	390	80	62	1
E2	Mo <sub>1</sub> V <sub>0.28</sub> Te <sub>0.13</sub> Nb <sub>0.13</sub> Pd <sub>0.001</sub>	0.73	420	77	59	1
CE3	Mo <sub>1</sub> V <sub>0.33</sub> Te <sub>0.22</sub> Nb <sub>0.11</sub> Pd <sub>0.04</sub>	0.35	440	75	42	1
E3	Mo <sub>1</sub> V <sub>0.29</sub> Te <sub>0.13</sub> Nb <sub>0.13</sub> Pd <sub>0.001</sub>	0.74	385	77	60	1
CE4	Mo <sub>1</sub> V <sub>0.33</sub> Te <sub>0.19</sub> Nb <sub>0.11</sub> Co <sub>0.005</sub>	0.24	440	79	44	1
E4	Mo <sub>1</sub> V <sub>0.29</sub> Te <sub>0.13</sub> Nb <sub>0.13</sub> Co <sub>0.004</sub>	0.73	390	76	62	2
CE5	Mo <sub>1</sub> V <sub>0.33</sub> Te <sub>0.19</sub> Nb <sub>0.11</sub> Cu <sub>0.01</sub>	0.27	420	59	56	3
E5	Mo <sub>1</sub> V <sub>0.28</sub> Te <sub>0.13</sub> Nb <sub>0.13</sub> Cu <sub>0.003</sub>	0.74	420	73	62	2
CE6	Mo <sub>1</sub> V <sub>0.33</sub> Te <sub>0.19</sub> Nb <sub>0.11</sub> Bi <sub>0.004</sub>	0.18	400	83	54	1
E6	Mo <sub>1</sub> V <sub>0.28</sub> Te <sub>0.15</sub> Nb <sub>0.14</sub> Bi <sub>0.005</sub>	0.70	410	77	62	1
CE7	Mo <sub>1</sub> V <sub>0.34</sub> Te <sub>0.18</sub> Nb <sub>0.11</sub> Pb <sub>0.004</sub>	0.30	440	78	43	1
E7	Mo <sub>1</sub> V <sub>0.28</sub> Te <sub>0.13</sub> Nb <sub>0.13</sub> Pb <sub>0.001</sub>	0.67	420	78	58	2
CE8	Mo <sub>1</sub> V <sub>0.33</sub> Te <sub>0.16</sub> Nb <sub>0.11</sub>	0.26	420	68	55	2
CE9	Mo <sub>1</sub> V <sub>0.29</sub> Te <sub>0.13</sub> Nb <sub>0.13</sub>	0.68	410	80	56	2

Comparative Example (CE1) and Inventive Example (E1) contain the same five metals (i.e., Mo, V, Te, Nb, and Ni), however the inventive example was prepared by a method that enriches the i-phase. The enriched i-phase nature of the inventive example is reflected in the column labeled R in the Table above.

The Comparative and Inventive Examples were tested for their ability to catalyze the oxidation of propane. The catalytic reactions of Comparative Example 1 and Inventive Example 1 are carried out under the same conditions (e.g., each is carried out at a temperature of 390°C). It is immediately evident from the results of the Table that the multimetal oxide material of the invention is able to provide a substantially higher propane conversion (i.e.,  $C_{PAN}$ ) and lower amounts of propene byproduct formation (i.e.,  $S_{PEN}$ ).

Comparative Example 5 (CE5) and Inventive Example 5 (E5) are carried out the same conditions (e.g., a temperature of 420°C). The inventive example adheres to the claim limitation that the i-phase is present in a greater quantity than the k-phase as indicated by the ratio R. The inventive example which is enriched in i-phase is able to provide substantially improved propane conversion (i.e.,  $C_{PAN}$  for the inventive example is 73 mol% whereas the comparative example is able to provide a  $C_{PAN}$  of only 59 mol%). There are likewise improvements in the propane by-product formation and in the selectivity of acrylic acid formation (i.e.,  $S_{ACA}$ ).

Applicants have therefore demonstrated that the multimetal oxide of the present invention which contains an i-phase crystal form of a 5-metal multimetal oxide is able to provide substantially improved propane conversion and selectivity of propene by-product formation in comparison to multimetal oxides that are not enriched in the i-phase.

Applicants have also compared 4-metal multimetal oxides with the 5-metal multimetal oxides of the claims. Comparative Example 9 contains a 4-metal (i.e. Mo, V, Te and Nb) multimetal oxide that adheres to the i-phase/k-phase requirement of present

independent Claim 1. The 4-metal multimetal oxide is able to provide good propane conversion (i.e.,  $C_{PAN} = 80$  mol%). However, the 4-metal multimetal oxide is able to provide only a low selectivity for acrylic acid formation (i.e., a  $S_{ACA}$  of 56 mol% in comparison to  $S_{ACA}$  for the 5-metal multimetal oxide of example E1 which is 66 mol%).

Applicants have therefore shown that the claimed 5-metal multimetal oxide is (i) distinguished from 4-metal multimetal oxides by improvements in the selectivity of propane to acrylic acid formation and (ii) that the 5-metal multimetal oxide provides significantly improved conversion and selectivity when it is enriched in the i-phase in comparison to 5-metal multimetal oxide materials manufactured using conventional techniques which provide a mixture of i- and k-phases.

WO 02/06199 is cited as a primary reference of the prior art references in the Office Action of May 10, 2005. The Office applies WO 02/06199 as disclosure of specific crystal structures that have a crystalline phase (i.e., the k phase). The following is disclosed with regard to WO 02/06199 at page 2, line 40 through page 3, line 8 of the present specification:

DE-A 19835247, EP-A 1090684 and **WO 02/06199** disclose that the abovementioned specific crystal structure forms only one crystalline phase in which such multimetal oxide materials can occur. This crystalline phase is referred to in the abovementioned literature as a rule as the k phase.

A second specific crystal structure in which the relevant multimetal oxide materials can occur is referred to as a rule as the i phase. Typical of its X-ray diffraction content, according to the abovementioned publications, is, inter alia, that it has

reflections of the strongest intensity at the  $2\Theta$  peak positions  $22.2 + 0.4^\circ$ ,  $27.3 + 0.4^\circ$  and  $28.2 + 0.4^\circ$ , in contrast to the k

phase in which, however, there is no reflection at the  $2\Theta$  peak position  $50.0 + 0.3^\circ$ .

The prior art multimetal oxide material is in a k-phase crystal form. As noted in the present application the k phase can be distinguished from the i phase by its peak positions and the intensity ratio R.

In comparison, JP-A 7-232071 and **WO 02/06199** disclose that multimetal oxide materials present exclusivity in the i-phase structure are also suitable as catalysts for the heterogeneously catalyzed partial ammoxidation and/or oxidation of saturated hydrocarbons.

Furthermore, experiments have already been carried out in which it is shown that multimetal oxide materials present exclusively in the k-phase structure are catalytically inactive and which support the concept of JP-A 11-169716 whereby the i phase is responsible for the activity and the k phase is responsible only for maximizing the selectivity.

Thus the present specification recognizes WO 02/06199 as prior art. However, WO 02/06199 shows only that 4-metal multimetal oxides may provide better catalytic activity when present in the i-phase in comparison to the k-phase. WO 02/06199 makes no mention that further improvements in performance may be obtained by using a 5-metal multimetal oxide composition in an i-phase. Applicants note that U.S. 2003/0187298 corresponds to WO 02/06199.

The improvements of WO 02/01699 are quantified (see the Table after paragraph [0158] in 2003/0187298). The best conversion obtained in the prior art is 28 mol% and the best selectivity is 72%. This may be compared with C<sub>PAN</sub> conversions of 73-80 mol% for the inventive examples described in the present specification. Thus Applicants have demonstrated that the conversion achieved in the claimed multimetal oxides may be more than 200% greater than that obtained with the WO 02/01699 multimetal oxides. Applicants note that even a 1 mol % increase in conversion and/or selectivity is significant when one considers that global demand for acrylic acid is more than one million metric tons.

How can a prior art multimetal oxide having a maximum disclosed conversion of only 28mol% render obvious a multimetal oxide having a conversion of as much as 80 mol%?

The Office admits that WO 02/01699 does not disclose a multimetal oxide of formula (I) of present Claim 1 because WO 02/01699 does not disclose a multimetal oxide having an M<sup>3</sup> component. The Office applies several secondary prior art references as evidence that it

would be obvious to include a third metal component (i.e.,  $M^3$  in formula (I)) to arrive at the claimed invention.

The Office applied Hinago (U.S. 6,610,629) to cure the defects of WO 02/01699. The Office admits that Hinago does not require the presence of a third metal atom such as  $M^3$  as required in formula (I) of present Claim 1. Hinago discloses a number of multimetal oxides in Tables 1 and 2. None of the Hinago multimetal oxides contain three metal atoms (i.e.,  $M^1$ ,  $M^2$ , and  $M^3$  of formula (I)). Applicants submit that Hinago does not suggest or disclose that the multimetal oxides of formula (I) of present Claim 1 can provide improved catalyst selectivity such as that evidenced by improved selectivity during acrylic acid preparation.

The Office further applies a series of patents to Chaturvedi (including U.S. 6,734,136; U.S. 6,407,280; U.S. 6,407,031; and U.S. 6,734,136). However, the Chaturvedi patents do not describe multimetal oxide compositions that are enriched in the i-phase. The production methods disclosed in Chaturvedi are conventional techniques that do not include any step of enriching, isolating or purifying the i-phase of the multimetal oxide. In fact, Chaturvedi teaches that the k-phase is a preferred phase. See for example the tables in columns 6 or 7 of the Chaturvedi patents which provide a description of the x-ray diffraction spectra of the Chaturvedi multimetal oxides and wherein each case a  $2\theta$  reflection at  $50.0^\circ$  is provided). As Applicants noted above, the  $50.0^\circ$  reflectance is indicative of the k-phase. Therefore, Chaturvedi is disclosing the use of the k-phase thereby teaching away from the i-phase enriched multimetal oxide composition of the present claims.

The Office characterizes the Chaturvedi disclosure as follows:

The reference teaches that the addition of IR and/or SM provides a catalyst with increased selectivity which results in greater yields of the desired reaction product (column 4, lines 15-22) (see page 4, lines 11-13 of the Office Action of May 10, 2005).



The disclosure of the Chaturvedi patent to which the Office cites is reproduced below for convenience:

By the present invention, there are provided promoted catalysts wherein the selectivity is greatly enhanced as to the base catalyst and, hence, the overall yield of the desired reaction product is also greatly enhanced.

The improvement that Chaturvedi shows for the “greatly enhanced” prior art catalysts is demonstrated in the tables of the patent (see Tables 1 and 2 in columns 17 and 18 of the 6,734,136 patent. The best propane conversion achieved by the prior art catalyst is disclosed in Table 1 as 48% (as already noted above the claimed invention is proven to provide a conversion of up to 80 mol%). Table 2 discloses a conversion of 76% however with a selectivity of only 13%.

How can the multimetal oxides of the claimed invention which are shown to provide high conversion and selectivity be obvious in view of prior art multimetal oxides that are nowhere disclosed to simultaneously provide high conversion and selectivity?

New dependent Claims 32 and 33 limit the conversion and selectivity of the claimed multimetal oxide under certain conditions.

New dependent Claims 30 and 31 have been added. New dependent Claim 30 requires that  $d$  in formula (I) is from 0.003 to 1. New dependent Claim 31 requires that  $d$  is from 0.004 to 1.

Applicants submit that a 5-metal multimetal oxide that is required to be enriched in the i-phase is not obvious in view of the 4- or 5-metal multimetal oxide k-phase compositions of the prior art.

Applicants submit that the presently claimed invention is not obvious in view of the prior art relied upon by the Office at least because of Applicants' showing that the i-phase 5-metal multimetal oxide compositions provide improved conversion and selectivity in comparison to 4-metal multimetal oxides or multimetal oxides that are not enriched in the i-

Application No. 10/684,559  
Reply to Office Action of May 10, 2005

phase. Applicants submit that Chaturvedi teaches away from the presently-claimed invention by favoring the k-phase. Applicants submit the presently claimed invention is further patentable over the prior art of record in view of Applicants' showing that 5-metal multimetal oxides are able to provide improved selectivity in comparison to 4-metal multimetal oxides. Importantly, Applicants have shown that the claimed multimetal oxides are able to concurrently provide high conversion and selectivity, a characteristic not disclosed for any of the prior art multimetal oxides.

Applicants submit the data of the specification are sufficient to rebut the Offices allegation of obviousness and respectfully request the withdrawal of the rejections.

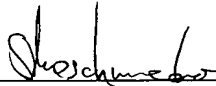
Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,  
MAIER & NEUSTADT, P.C.  
Norman F. Oblon

Customer Number

**22850**

Tel: (703) 413-3000  
Fax: (703) 413 -2220  
(OSMMN 06/04)



---

Stefan U. Koschmieder, Ph.D.  
Registration No. 50,238

NFO:SUK\la